Electronic Supplementary Information for *Soft Matter* Manuscript:

Formation and Control of Highly Crumpled Metal Surfaces on Photocurable Viscous Liquid

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Figures in Supplementary Information

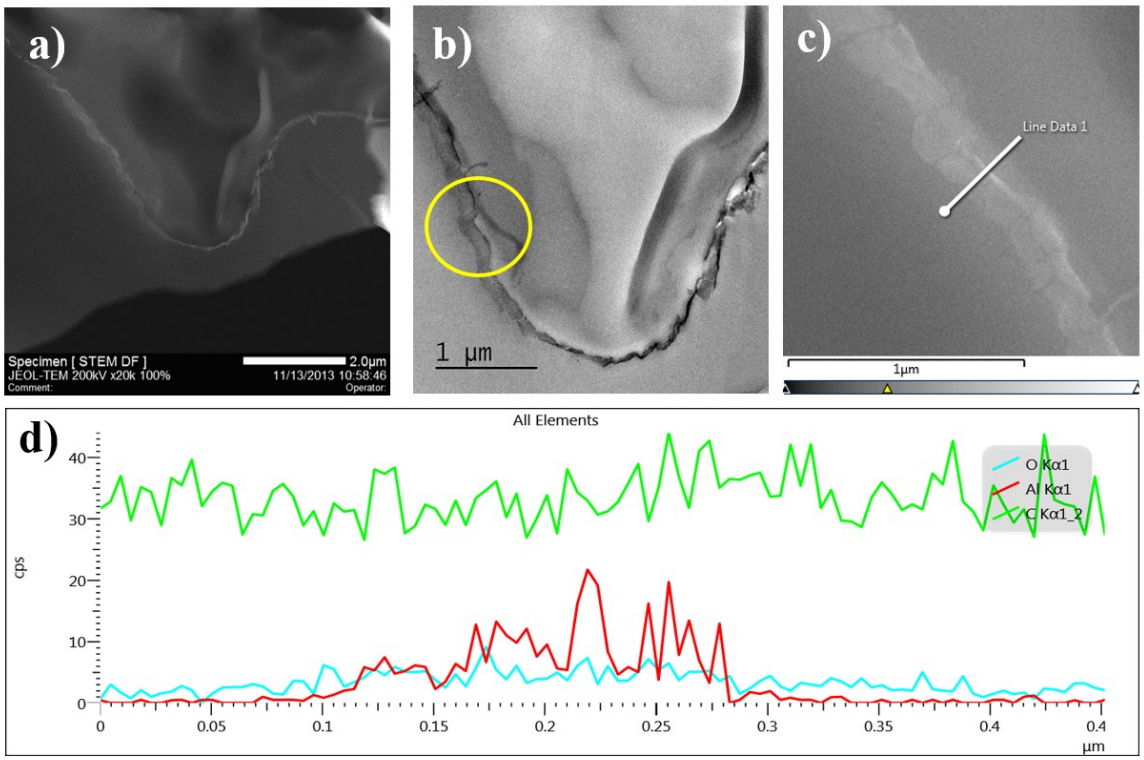

Fig. S1 a) A cross-sectional SEM image of deformed surface. b) A cross-sectional TEM image of the sliced sample prepared by microtoming. The black lines represent aluminum metal film and the bright region is polymers. The grey region in the yellow circle is the oxidized aluminum. c) A SEM image showing the region to analyze with Energy-dispersive X-ray spectroscopy (EDX). d) A EDX result represent the aluminum and oxygen is located on the film surface.

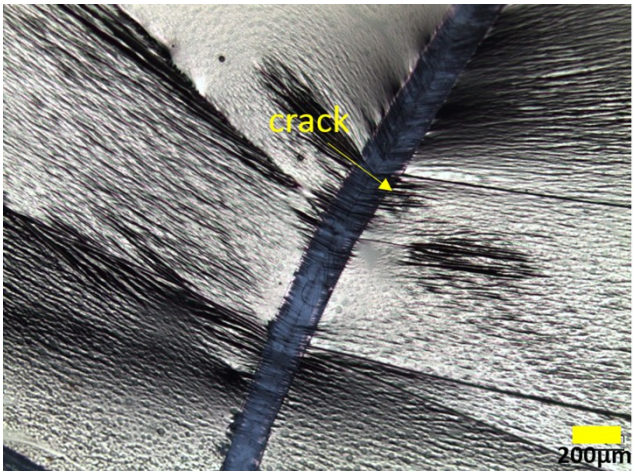

Fig. S2 Optical microscope image of cracked surface via Au deposition (10 nm thick, 0.5 Å/sec) on an un-cured viscous prepolymer. When Au thin film is deposited onto the prepolymer substrate, there exists a tensile stress. Then crack formation occurs.

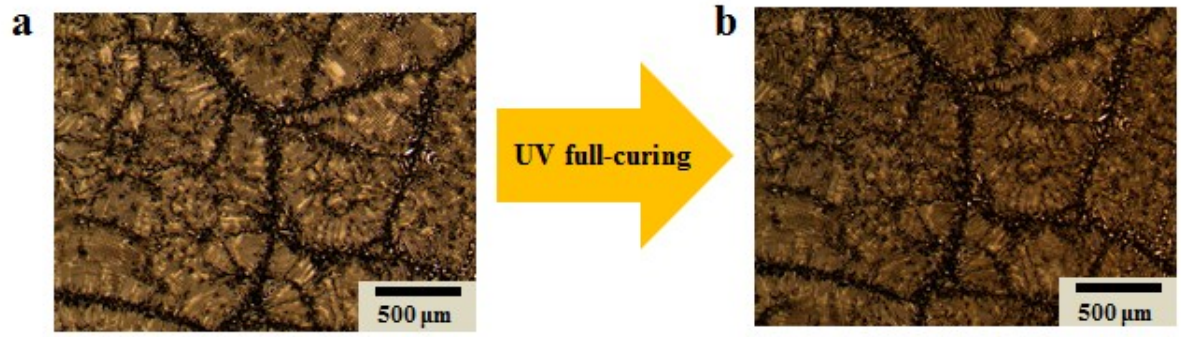

Fig. S3 Optical microscope images of crumpled thin metal film a) before and b) post UV exposure. There is little difference between them. The deformation of metal film is irreversible; thus the shape of the film is maintained during post crosslinking process.

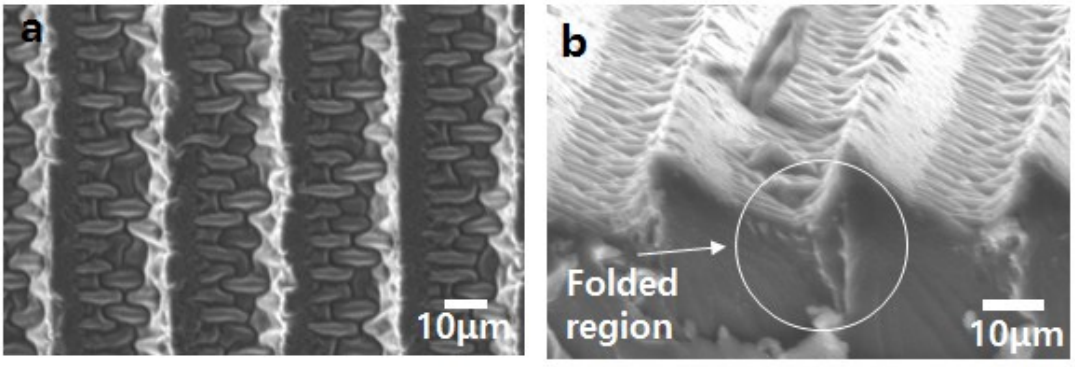

Fig. S4 Planar a) and cross-sectional b) SEM images of the deformed surface during the deposition of metal films onto a viscous liquid confined by an asymmetric ratchet array whose period and depth are both 30 μm. The viscous liquid is coated by doctor blading method.

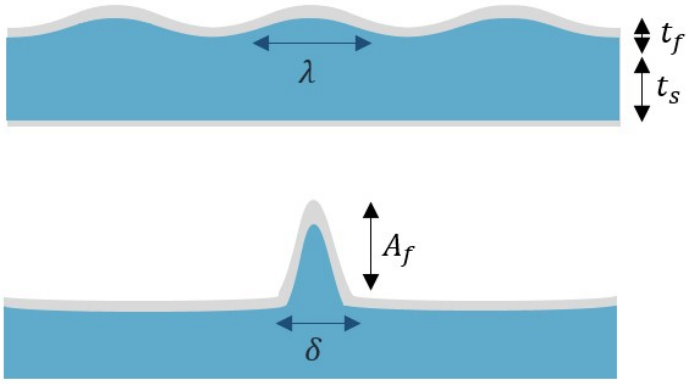

Fig. S5 Schematic illustration for the theoretical consideration of relation between amplitude of folding and thickness of substrate.

Scaling analysis for folding amplitude

To explain the relationship between folding amplitude and polymer thickness, a simple scaling theory is developed. When the system is under biaxial stress, it can be modeled as a 1D problem based on the assumption that the folding formation mechanism is the same in both directions.

According to Cerda and Mahadevan, the wavelength is represented by the scaling relation, $\lambda \sim (B/K)^{1/4}$, where B is a bending modulus and K is the effective stiffness of the substrate. When the thickness of the substrate t_s is smaller than the characteristic length scale of the buckling, the stiffness of the substrate is expressed as $K \sim E_s\lambda^2/{t_s}^3$ where E_s is the Young's modulus of the substrate.[1]

Assuming that the time scale of the increase of compressive stress is much shorter than the relaxation time scale of the polymer substrate and prepolymer substrate has linear viscoelasticity, the modulus of the polymer substrate can be approximated to be the shear modulus G, which is independent of time. There is also a linear relationship between G and E_s given by $G = E_s/(2(1+\nu))$, where ν is Poisson's ratio.[2] Then, stiffness of polymer substrate is expressed as $K \sim G\lambda^2/t_s^3$.

Substituting the equation of the wavelength $\lambda \sim (B/K)^{1/4}$ where bending stiffness $B \sim E_f{t_f}^3$, then $\lambda \sim ((E_f/G)(t_f t_s)^3\lambda^{-2})^{1/4}$ where E_f and t_f are Young's modulus and the thickness of thin film, respectively. As a result, the wavelength of the system is given by the following equation: $\lambda \sim (E_f/G)^{1/6}(t_f t_s)^{1/2}$

Because all the experimental parameters except the thickness of substrate are kept constant in our experiments, the wavelength has a scaling relation of $\lambda \sim (t_s)^{1/2}$. The bending energy has a form of $U_B = \frac{B}{2}\int_0^L (\frac{d^2y}{dx^2})^2 dx$ where L is a length of the film. That scale as

$U_{B,w} \sim BL(A/\lambda^2)^2 \sim B\Delta_b/\lambda^2$ and $U_{B,f} \sim B/\delta$, buckling and folding respectively where Δ_b is a horizontal deformation in buckling system and δ is a width of the folding.[3] Comparing these two energies, they are nearly equivalent at the buckling to folding transition point, which results in $\Delta_b \sim \lambda^2/\delta$. In this critical region, ratio of Δ_b and Δ_f is nearly unity, where Δ_f is horizontal deformation of folding system. Folding grows from one of peak of the bucklings, δ is proportional to λ. Therefore, deformation for folding scales as $\Delta_f \sim \lambda$ (see Fig. S5).

In addition, the amplitude of folding can be approximated to $\Delta/2$ after increasing the strain where the curvature localization is perfect. By using these relations, a final formula related to the amplitude of folding A_f is $A_f \sim {t_s}^{1/2}$.

1 E. Cerda and L. Mahadevan, *Phys. Rev. Lett.*, 2003, **90**, 74302.
2 J. Gere and B. Goodno, *Mechanics of materials*, Nelson Education, 2012.
3 L. Pocivavsek, R. Dellsy, A. Kern, S. Johnson, B. Lin, K. Y. C. Lee and E. Cerda, *Science (80-.).*, 2008, **320**, 912–916.

Video Captions in Supplementary Information

Video S1. Real time movie for purging of nitrogen gas on the deformed metal thin film without UV exposure. The metal thin film was pulverized due to applied pressure.

Video S2. Real time movie for purging of nitrogen gas on the deformed metal thin film after UV exposure. It is verified that, after UV exposure, the film is well attached on the polymer substrate.